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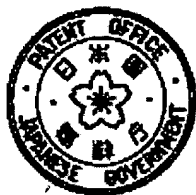
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(84) Designated contracting states:	(74) Representative:

**(54) POLYETHYLENE OXIDE  
GROUP-CONTAINING  
POLYSILOXANE CROSS-  
LINKED PRODUCT AND  
POLYMER SOLID  
ELECTROLYTE**

(57) Abstract:

**PROBLEM TO BE SOLVED:** To provide a polymer solid electrolyte which has high ion conductivity even in a solvent-free system and has excellent mechanical strengths.

**SOLUTION:** In the polysiloxane cross-linked product, structural unit comprising a tri-functional silicon-having polysiloxane are bound to structural units comprising a compound having oxyethylene units as repeating units. The polymer solid electrolyte comprises the polysiloxane cross-linked product and a lithium salt.

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**Title:** JP2003002974A2: POLYETHYLENE OXIDE GROUP-CONTAINING POLYSILOXANE CROSS- LINKED PRODUCT AND POLYMER SOLID ELECTROLYTE

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YOZA AKIRA

OIKAWA HISAO

YAMAMOTO YASUHIRO

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**(54) POLYETHYLENE OXIDE GROUP-CONTAINING POLYSILOXANE CROSS- LINKED  
PRODUCT AND POLYMER SOLID ELECTROLYTE**

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(54) 【発明の名称】 ポリエチレンオキシド基含有ポリシロキサン架橋体および高分子固体電解質

(57) 【要約】

【課題】 無溶媒系においても高いイオン伝導度を有し、しかも機械的強度に優れた高分子固体電解質を提供すること。

【解決手段】 3官能性ケイ素を有するポリシロキサンからなる構造単位と、オキシエチレンを繰り返し単位として有する化合物からなる構造単位とが結合されていることを特徴とするポリシロキサン架橋体、およびこのポリシロキサン架橋体とリチウム塩とからなる高分子固体電解質。

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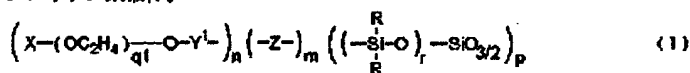
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## 【特許請求の範囲】

【請求項1】3官能性ケイ素を有するポリシロキサンからなる構造単位と、オキシエチレンを繰り返し単位として有する化合物からなる構造単位とが結合されていることを特徴とするポリシロキサン架橋体。 \*



(式中、Xは炭素数1～6のアルキルであり、Y<sup>1</sup>は炭素数2～20のアルキレンであり、Zは両末端に炭素数2～20のアルキレンを有する基であり、Rは炭素数1～6のアルキルであり、q1、nおよびmはそれぞれ独立して1以上の整数、rは0または1、pは4～60の※



(式中、Y<sup>2</sup>およびY<sup>3</sup>はそれぞれ独立して炭素数2～20のアルキレン、q2は1以上の整数である。)

【請求項4】q2が3～15であることを特徴とする、請求項3に記載のポリシロキサン架橋体。

【請求項5】nとmの関係が、2m/(n+2m)が0.2～0.6であるように調整されていることを特徴とする、請求項2～4のいずれか1項に記載のポリシロキサン架橋体。

【請求項6】rが0であることを特徴とする、請求項2～5のいずれか1項に記載のポリシロキサン架橋体。

【請求項7】rが1であることを特徴とする、請求項2～5のいずれか1項に記載のポリシロキサン架橋体。

【請求項8】請求項1～7のいずれか1項に記載のポリシロキサン架橋体とリチウム塩とからなる高分子固体電解質。

## 【発明の詳細な説明】

## 【0001】

【発明の属する技術分野】本発明は一次電池、二次電池、コンデンサー等に使用できる高分子固体電解質であって、3官能性ケイ素を有するポリシロキサンとポリオキシエチレンユニットとの組み合わせることにより得られるポリシロキサン架橋体、およびこれを用いて得られる、高いイオン伝導性と良好な機械的強度を持つ高分子固体電解質に関する。

## 【0002】

【従来の技術】高分子固体電解質は、電池の薄型化、無漏液化、耐熱性の向上を実現する材料として注目を集めている。高分子固体電解質には、イオン伝導度が高い、機械的強度が強い、電気化学的に安定、熱的に安定などの特性が要求されており、従来、これらの特性を満足する材料として、ガラス転移温度が低いことによるポリマーの運動性の高さ及びリチウムイオン溶解性の点でポリエチレンオキッド等のエーテル系ポリマーが有望視されている。また、イオンを効率良く動かすための手段としては、ガラス転移温度の低いポリマーにグラフト化する

\*【請求項2】ポリシロキサン架橋体が式(1)で表される組成の重合体であることを特徴とする、請求項1に記載のポリシロキサン架橋体。

## 【化1】

※整数であって、n+2m=pである。)

10 【請求項3】Zが式(2)で表される基であることを特徴とする、請求項2に記載のポリシロキサン架橋体。

## 【化2】

方法が多く用いられている。

【0003】特に、ポリシロキサンを主鎖に用いた高分子固体電解質は、ガラス転移温度が低いことからエーテル系ポリマーとの組み合わせによる研究が盛んになされている。さらに、ポリシロキサンの骨格をなすシロキサン結合はその結合エネルギーが炭素-炭素結合よりかなり大きく電気化学的に安定であるために、高分子固体電解質の材料として有用である。特開平4-56002号公報にポリシロキサンとポリエーテルグリコールとポリエーテルグリコール-モノメチルエーテルのグラフト共重合体からなる高分子固体電解質が開示され、特開平7-233243号公報にカルボキシル基含有炭化水素基を有するオルガノポリシロキサンとエポキシ基を有するポリオキシアルキレンとの共重合体からなる高分子固体電解質が開示されている。

## 【0004】

【発明が解決しようとする課題】しかしながら、ポリシロキサンを用いたいずれの場合であっても、充分なイオン伝導度は得られず実用化には至っていない。しかもこの方法を用いてイオン伝導率の向上を図ると、逆に機械的強度や成膜性を低下させるという問題点を抱えている。本発明は、無溶媒系においても高いイオン伝導度を有し、さらに優れた機械的強度を持ち、電気化学的にも熱的にも安定である高分子固体電解質を提供することを目的とする。

## 【0005】

【課題を解決するための手段】本発明者らは、シロキサンに着目して研究を重ねた結果、3官能性ケイ素を有するポリシロキサンとポリオキシエチレンユニットを有する化合物とからなるポリシロキサン架橋体を用いることにより、シロキサン結合の安定性を保ちつつ大幅に機械的特性が向上し、しかも無溶媒系におけるイオン伝導度が、直鎖のポリシロキサンとポリオキシエチレンユニットを有する有機高分子とのグラフト共重合体と同等以上である高分子固体電解質が得られることを見出し、本発



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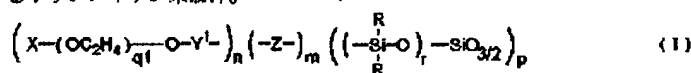
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明を達成した。即ち、本発明は下記の構成を有する。

【0006】(1) 3官能性ケイ素を有するポリシロキサンからなる構造単位と、オキシエチレンを繰り返し単位として有する化合物からなる構造単位とが結合されていることを特徴とするポリシロキサン架橋体。

\* (2) ポリシロキサン架橋体が式(1)で表される組成の重合体であることを特徴とする、前記(1)項に記載のポリシロキサン架橋体。

【化3】



(式中、Xは炭素数1～6のアルキルであり、Y<sup>1</sup>は炭素数2～20のアルキレンであり、Zは両末端に炭素数2～20のアルキレンを有する基であり、Rは炭素数1～6のアルキルであり、q1、nおよびmはそれぞれ独立して1以上の整数、rは0または1、pは4～60の※

※整数であって、n+2m=pである。)

(3) Zが式(2)で表される基であることを特徴とする、前記(2)項に記載のポリシロキサン架橋体。

【化4】



(式中、Y<sup>2</sup>およびY<sup>3</sup>はそれぞれ独立して炭素数2～20のアルキレン、q2は1以上の整数である。)

(4) q2が3～15であることを特徴とする、前記(3)項に記載のポリシロキサン架橋体。

(5) nとmの関係が、2m/(n+2m)が0.2～0.6であるように調整されていることを特徴とする、前記(2)～(4)項のいずれか1項に記載のポリシロキサン架橋体。

(6) rが0であることを特徴とする、前記(2)～(5)項のいずれか1項に記載のポリシロキサン架橋体。

(7) rが1であることを特徴とする、前記(2)～(5)項のいずれか1項に記載のポリシロキサン架橋体。

(8) 前記(1)～(7)項のいずれか1項に記載のポリシロキサン架橋体とリチウム塩とからなる高分子固体電解質。

【0007】

【発明の実施の形態】本発明におけるポリシロキサン架橋体は、3官能性ケイ素を有するポリシロキサンと、オキシエチレンを繰り返し単位として有する化合物とを結合させてなるポリシロキサン架橋体である。このポリシロキサン架橋体の合成法としては、例えば、3官能性ケイ素を有し、末端にSi-H基を有するポリシロキサン、両末端に二重結合を有する化合物、および片末端に二重結合を有しポリオキシエチレンユニットを含む化合物を、これらの化合物を溶解可能な溶媒中で混合し、これに触媒を投入してヒドロシリル化反応により重合させる方法がある。

【0008】3官能性ケイ素を有し、Si-H基を有するポリシロキサンは、市販されている製品を使用してもよく、また従来公知であるいかなる手段を用いて合成してもよい。よく知られている例としては、一般式Si<sub>3</sub>zH<sub>2z+2</sub>O<sub>z</sub>。(nが4または5のものが知られている。)で表されるシルセスキオキサンがあり、またこれ

らの化合物のSi-H基の一部または全部をSi-O-SiR<sub>2</sub>-H基(Rは、炭素数1～6のアルキル基を示す。)で置き換えた化合物を挙げることができる。

【0009】次に、両末端に二重結合を有する化合物は、3官能性ケイ素を有するポリシロキサン同士を結合するための連結基として働くものであり、例えば、ポリエチレングリコールジアリルエーテルや末端ビニルポリジメチルシロキサンが挙げられる。また、片末端に二重結合を有しポリオキシエチレンユニットを含む化合物としては、例えば、トリエチレングリコールアリルメチルエーテル、トリエチレングリコールメチルビニルエーテル、トリエチレングリコールアリルエチルエーテル、ポリエチレングリコールアリルメチルエーテルなどが挙げられる。

【0010】ヒドロシリル化反応に用いられる有機溶媒としては、例えば、トルエン、テトラヒドロフラン(THF)などが挙げられる。また、ヒドロシリル化触媒としては、例えば、白金化合物、ルテニウム化合物、ロジウム化合物が挙げられる。

【0011】本発明の高分子固体電解質は、上記のポリシロキサン架橋体にリチウム塩を配合したものである。リチウム塩の配合方法は特に限定されるものではないが、例えば、ポリシロキサン架橋体とリチウム塩を有機溶媒中で均一に混合し、その後、減圧、加熱下で有機溶媒を完全に除去する方法が挙げられる。用いられる有機溶媒としては、リチウム塩を溶解可能な限り限定されないが、例えば、THF、ジメチルホルムアミド、アセトンなどが挙げられる。

【0012】リチウム塩としては、高分子固体電解質に通常用いられるものであればいかなるリチウム塩であってもよく、例えば、LiF、LiCl、LiBr、LiI、LiClO<sub>4</sub>、LiBF<sub>4</sub>、LiPF<sub>6</sub>、LiAsF<sub>6</sub>、LiCF<sub>3</sub>SO<sub>3</sub>、LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>、LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>、およびLiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>、等を挙げることができ、これらから少なくとも1

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種以上のリチウム塩を選んで用いればよい。

【0013】

【実施例】本発明を実施例に基づいてさらに詳細に説明するが、本発明はそれら実施例に限定されるものではない。

合成例1

<ポリシロキサン架橋体の合成1>100mL四つ口フラスコに、モレキュラーシーブ3Aで1晩以上脱水したトルエン15mL、トリエチレングリコールアリルメチルエーテル0.803g、平均分子量300のポリエチレングリコールジアルキルエーテル0.748g、および1,3,5,7,9,11,13,15-オクタキス(ジメチルシロキシ)ペンタシクロ[9.5.1.1<sup>3,5</sup>.1<sup>7,9</sup>.1<sup>11,13</sup>]オクタシロキサン1.00gを入れ、窒素シールド下、80℃で1時間加熱攪拌した。次いで、白金ジビニルテトラメチルジシロキサンをSi-Hに対し50ppm投入し、そのまま3時間加熱攪拌を続けた。IRでSi-Hのピークが消失していることを確認した後、400Paの減圧下、120℃で低沸分を除去して、褐色透明固体のポリシロキサン架橋体をほぼ定量的に得た。得られたポリシロキサン架橋体をPS1とする。

【0014】実施例1

<固体電解質および測定用セルの作成、イオン伝導度の測定>合成例1で得られたPS1の0.1gに対して、[リチウムイオン]/[オキシエチレンユニット]が当量比0.05になるように、LiClO<sub>4</sub> 5.98×10<sup>-3</sup>gをTHF中で均一に混合した後、400Paの\*

$$(\text{弾性率}) = \{ (\text{荷重振幅}) / (\text{プローブ先端面積}) \} / \{ (\text{TMA振幅}) / (\text{サンプルの初期長さ}) \} \quad (\text{定義式1})$$

【0016】比較合成例1

<ポリシロキサングラフト架橋体の合成>200mL四つ口フラスコに、モレキュラーシーブ3Aで1晩以上脱水したトルエン50mL、トリエチレングリコールアリルメチルエーテル16.92g、トリエチレングリコールジアルキルエーテル4.14g、白金ジビニルテトラメチルジシロキサンを、平均分子量1700のポリメチルヒドロシロキサン5.00g中のSi-Hに対し50ppmになるように入れ、窒素シールド下、80℃で1時間加熱攪拌した。次いで、前記のポリメチルヒドロシロキサン5.00gを脱水トルエン10mLで希釈した溶液を滴下した。そのまま1時間加熱攪拌を続けて、IRでSi-Hのピークが消失していることを確認した後、400Paの減圧下、130℃で低沸分を除去して、褐色透明固体のポリシロキサングラフト架橋体をほぼ定量的に得た。

【0017】比較例1

<固体電解質および測定用セルの作成、イオン伝導度の測定>上記のポリシロキサングラフト架橋体0.2gに対して、[リチウムイオン]/[オキシエチレンユニッ

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\*減圧下、100℃で3時間加熱し、THFを完全に除去して固体電解質を作成した。この固体電解質をステンレス電極ではさんで熱プレスにより成形し、冷却後、これを用いて測定用セルを作成した。作成した測定用セルを周波数応答分析計(ソーラトロン社製S1-1287)を用い、複素インピーダンス測定法(交流振幅電圧1V、交流の周波数帯域1Hz~2MHz、温度30℃)によりインピーダンスを測定し、イオン伝導度を算出した。結果を表1に示した。

10 【0015】実施例2

<弾性率測定用フィルムの作成および測定>合成例1と全く同様にしてポリシロキサングラフト架橋体を合成し、反応終了後に溶液の一部を直径30mmのポリフッ化エチレン製シャーレに分取した。これを、気泡が入らないよう徐々に減圧しながら低沸分を除去した。更に400Paの減圧下、130℃で乾燥し、測定用のフィルムを作成した。作成したフィルムから肉厚2mm×直径5mmの試験迎を切り出し、セイコー電子工業(株)製の熱機械的分析装置TMA100を用いて、以下の①~③の手順に従い、室温における弾性率の測定を行った。①TMA100に作成したフィルムをセットし、荷重98mNを加えた状態でサンプルの初期長さを測定する。②98mN±19.6mNの荷重振幅でのTMA振幅を測定する(周波数0.01Hz)。③弾性率の定義式(定義式1)に、プローブ先端面積(5.3mm<sup>2</sup>)、荷重振幅(39.2mN)、サンプルの初期長さ、TMA振幅を代入し、弾性率を算出する。得られた弾性率を表2に示した。

ト)が当量比0.05になるように、0.0115gのLiClO<sub>4</sub>をTHF中で均一に混合した後、400Paの減圧下、100℃で3時間加熱し、THFを完全に除去して固体電解質を作成した。この固体電解質をステンレス電極ではさんで熱プレスにより成形し、冷却後、これを用いて測定用セルを作成した。作成した測定用セルを周波数応答分析計(ソーラトロン社製S1-1287)を用い、複素インピーダンス測定法(交流振幅電圧1V、交流の周波数帯域1Hz~2MHz、温度30℃)によりインピーダンスを測定し、イオン伝導度を算出した。結果を表1に示した。

【0018】比較例2

<弾性率測定用フィルムの作成および測定>比較合成例1と全く同様にしてポリシロキサングラフト架橋体を合成し、反応終了後に溶液の一部を直径30mmのポリフッ化エチレン製シャーレに分取した。これを、気泡が入らないよう徐々に減圧しながら低沸分を除去した。更に400Paの減圧下、130℃で乾燥し、測定用のフィルムを作成した。作成したフィルムから肉厚2mm×直径5mmの試験迎を切り出し、セイコー電子工業(株)

(5)

特開2003-2974

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製の熱機械的分析装置TMA100を用いて、実施例2 \* [0019]  
 と同様の手順で、室温における弾性率の測定を行った。【表1】  
 得られた弾性率を表2に示した。 \*

&lt;表1&gt;

	実施例1	比較例1
イオン伝導度(S/cm)	$1.8 \times 10^{-5}$	$1.8 \times 10^{-5}$

【表2】

&lt;表2&gt;

	実施例2	比較例2
弾性率(MPa)	0.53	0.20

上記表1および表2に示した結果から、実施例のポリシロキサン架橋体は、比較例のポリシロキサン架橋体に比べて、イオン伝導度を下げることなく良好な機械的強度を有していることが分かる。

[0020]

\*

※【発明の効果】本発明により、無溶媒系においても高いイオン伝導度を有し、さらに優れた機械的強度を持ち、電気化学的にも熱的にも安定である高分子固体電解質を得ることが可能となった。

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**CLAIMS**

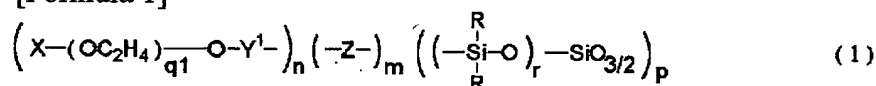

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[Claim(s)]

[Claim 1] The polysiloxane bridge formation object characterized by combining the structural unit which consists of a polysiloxane which has 3 functionality silicon, and the structural unit which consists of a compound which repeats oxyethylene and it has as a unit.

[Claim 2] The polysiloxane bridge formation object according to claim 1 characterized by being the polymer of the composition as which a polysiloxane bridge formation object is expressed in a formula (1).

[Formula 1]



(X is the alkyl of carbon numbers 1-6 among a formula, Y1 is the alkylene of carbon numbers 2-20, Z is a basis which has the alkylene of carbon numbers 2-20 in both ends, R is the alkyl of carbon numbers 1-6, 0 or 1p of one or more integers and r is the integer of 4-60, and q1, n, and m are n+2 m=p independently, respectively.)

[Claim 3] The polysiloxane bridge formation object according to claim 2 characterized by Z being the basis expressed with a formula (2).

[Formula 2]



(As for Y2 and Y3, the alkylene of carbon numbers 2-20 and q2 are one or more integers independently among a formula, respectively.)

[Claim 4] The polysiloxane bridge formation object according to claim 3 characterized by q2 being 3-15.

[Claim 5] A polysiloxane bridge formation object given in any 1 term of claims 2-4 characterized by adjusting the relation between n and m so that 2m/(n+2m) may be 0.2-0.6.

[Claim 6] A polysiloxane bridge formation object given in any 1 term of claims 2-5 characterized by r being 0.

[Claim 7] A polysiloxane bridge formation object given in any 1 term of claims 2-5 characterized by r being 1.

[Claim 8] The solid polymer electrolyte which becomes any 1 term of claims 1-7 from the polysiloxane bridge formation object and lithium salt of a publication.

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention is the solid polymer electrolyte which can be used for a primary cell, a rechargeable battery, a capacitor, etc., and relates to the high ion conductivity and the solid polymer electrolyte with a good mechanical strength which are obtained using the polysiloxane bridge formation object acquired by [ of the polysiloxane and polyoxyethylene unit which have 3 functionality silicon ] combining, and this.

[0002]

[Description of the Prior Art] The solid polymer electrolyte attracts attention as a material which realizes thin-shape-izing of a cell, spill liquid[ no ]-izing, and heat-resistant improvement. Which stable property is demanded stably and thermally electrochemically, and promising \*\* of the ether system polymer, such as a polyethylene oxide, is conventionally carried out in respect of the motile height of the polymer by a glass transition temperature being low, and lithium ion solubility as a material with a mechanical strength strong against a solid polymer electrolyte with high ionic conductivity with which are satisfied of these properties. Moreover, many methods of graft-izing are used for the low polymer of a glass transition temperature as a means for moving ion efficiently.

[0003] Since the glass transition temperature is low, especially as for the solid polymer electrolyte which used the polysiloxane for the principal chain, research by combination with ether system polymer is made briskly. Furthermore, since the binding energy is quite larger than a carbon-carbon to carbon bond and siloxane combination which makes the frame of a polysiloxane has it, it is useful as a material of a solid polymer electrolyte. [ electrochemically stable ] The solid polymer electrolyte which becomes JP,4-56002,A from the graft copolymer of a polysiloxane, a polyether glycol, and the polyether glycol monomethyl ether is indicated, and the solid polymer electrolyte which consists of a copolymer of the organopolysiloxane which has a carboxyl group content hydrocarbon group in JP,7-233243,A, and the polyoxyalkylene which has an epoxy group is indicated.

[0004]

[Problem(s) to be Solved by the Invention] However, even if it is which [ using the polysiloxane ] case, sufficient ionic conductivity is not obtained and has not resulted in utilization. And if improvement in ionic conductivity is aimed at using this method, the trouble of reducing a mechanical strength and membrane formation nature conversely is held. this invention has high ionic conductivity also in a non-solvent system, has the further excellent mechanical strength, and aims at offering a stable solid polymer electrolyte also electrochemically and thermally.

[0005]

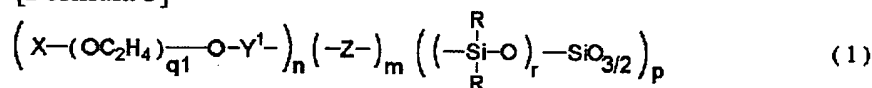
[Means for Solving the Problem] This invention persons by using the polysiloxane bridge formation object which consists of a polysiloxane which has 3 functionality silicon, and a compound which has a polyoxyethylene unit as a result of repeating research paying attention to a siloxane A mechanical property improves sharply, maintaining the stability of siloxane combination, and moreover the ionic conductivity in a non-solvent system It found out that the graft copolymer of the polysiloxane of a straight chain and the organic macromolecule which has a polyoxyethylene unit, and the solid polymer electrolyte which is more than equivalent were obtained, and this invention was attained. That is, this invention has the following composition.

[0006] (1) The polysiloxane bridge formation object characterized by combining the structural unit

which consists of a polysiloxane which has 3 functionality silicon, and the structural unit which consists of a compound which repeats oxyethylene and it has as a unit.

(2) A polysiloxane bridge formation object given in the aforementioned (1) term characterized by being the polymer of the composition as which a polysiloxane bridge formation object is expressed in a formula (1).

[Formula 3]



(X is the alkyl of carbon numbers 1-6 among a formula, Y1 is the alkylene of carbon numbers 2-20, Z is a basis which has the alkylene of carbon numbers 2-20 in both ends, R is the alkyl of carbon numbers 1-6, 0 or 1p of one or more integers and r is the integer of 4-60, and q1, n, and m are n+2 m=p independently, respectively.)

(3) A polysiloxane bridge formation object given in the aforementioned (2) term characterized by Z being the basis expressed with a formula (2).

[Formula 4]



(As for Y2 and Y3, the alkylene of carbon numbers 2-20 and q2 are one or more integers independently among a formula, respectively.)

(4) A polysiloxane bridge formation object given in the aforementioned (3) term characterized by q2 being 3-15.

(5) A polysiloxane bridge formation object given in any 1 term of the aforementioned (2) - (4) term characterized by adjusting the relation between n and m so that  $2m/(n+2m)$  may be 0.2-0.6.

(6) A polysiloxane bridge formation object given in any 1 term of the aforementioned (2) - (5) term characterized by r being 0.

(7) A polysiloxane bridge formation object given in any 1 term of the aforementioned (2) - (5) term characterized by r being 1.

(8) The solid polymer electrolyte which becomes any 1 term of the aforementioned (1) - (7) term from the polysiloxane bridge formation object and lithium salt of a publication.

[0007]

[Embodiments of the Invention] The polysiloxane bridge formation object in this invention is a polysiloxane bridge formation object which makes it come to join together the polysiloxane which has 3 functionality silicon, and the compound which repeats oxyethylene and it has as a unit. The polysiloxane which has 3 functionality silicon and has an Si-H basis at the end as a synthesis method of this polysiloxane bridge formation object, for example, the compound which has a double bond in both ends, and the compound which has a double bond at the piece end and contains a polyoxyethylene unit in it are mixed in the solvent which can dissolve these compounds, and there is the method of supplying a catalyst to this and carrying out a polyaddition by the hydrosilylation reaction.

[0008] The product marketed may be used for the polysiloxane which has 3 functionality silicon and has an Si-H basis, and it may compound it using what conventionally well-known means. The compound which there is silsesquioxane expressed with general formula  $\text{Si}_2\text{nH}_2\text{nO}_3\text{n}$  (the thing of 4 or 5 is known for n.) as an example known well, and replaced a part or all of an Si-H basis of these compounds by 2-H Si-O-SiR (R shows the alkyl group of carbon numbers 1-6.) can be mentioned.

[0009] Next, the compound which has a double bond in both ends works as a connection machine for combining the polysiloxanes which have 3 functionality silicon, and polyethylene-glycol diaryl ether and end vinyl poly dimethylsiloxane are mentioned. Moreover, as a compound which has a double bond at the piece end and contains a polyoxyethylene unit in it, a triethylene-glycol allyl-compound methyl ether, the triethylene-glycol methyl vinyl ether, triethylene-glycol allyl-compound ethyl ether, a polyethylene-glycol allyl-compound methyl ether, etc. are mentioned, for example.

[0010] As an organic solvent used for a hydrosilylation reaction, toluene, a tetrahydrofuran (THF),

etc. are mentioned, for example. Moreover, as a hydrosilylation catalyst, a platinum compound, a ruthenium compound, and a rhodium compound are mentioned, for example.

[0011] The solid polymer electrolyte of this invention blends lithium salt with the above-mentioned polysiloxane bridge formation object. Although especially the combination method of lithium salt is not limited, lithium salt is uniformly mixed with a polysiloxane bridge formation object in an organic solvent, and the method of removing an organic solvent completely under reduced pressure and heating is mentioned after that, for example. Although it will not be limited as an organic solvent used especially if it is dissolution possible about lithium salt, THF, a dimethylformamide, an acetone, etc. are mentioned, for example.

[0012] As lithium salt, as long as it is usually used for a solid polymer electrolyte, you may be what lithium salt. For example, LiF, LiCl, LiBr, LiI, LiClO<sub>4</sub>, LiBF<sub>4</sub>, LiPF<sub>6</sub> and LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> and LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, and LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub> grade can be mentioned. What is necessary is to choose at least one or more sorts of lithium salt from these, and just to use.

[0013]

[Example] Although this invention is further explained to a detail based on an example, this invention is not limited to these examples.

Toluene 15mL [ which dehydrated one or more evenings by molecular-sieve 3A in the four synthetic example 1 <composition 1 of polysiloxane bridge formation object> 100mL(s) mouth flask ], and triethylene-glycol allyl-compound methyl-ether 0.803g, polyethylene-glycol diaryl ether 0.748g of average molecular weight 300 and 1, 3, 5, 7, 9, 11 and 13, and 15-OKUTAKISU (dimethyl siloxy) PENTA cyclo [9. 5.1.13, 9.15, 15.17, and 13] OKUTA siloxane 1.00g. It put in and heating stirring was carried out at 80 degrees C under the N<sub>2</sub> seal for 1 hour. Subsequently, 50 ppm of platinum divinyl tetramethyl disiloxane were supplied to Si-H, and heating stirring was continued as it was for 3 hours. After checking that the peak of Si-H has disappeared by IR, the low-boiling part was removed at 120 degrees C under reduced pressure of 400Pa, and the polysiloxane bridge formation object of a brown transparent solid-state was acquired almost quantitatively. The acquired polysiloxane bridge formation object is set to PS1.

[0014] After mixing uniformly 45.98x10 to 3 g LiClO in THF to 0.1g of PS1 obtained in the example 1 of example 1 <measurement of creation [ of a solid electrolyte and the cell for measurement ], and ionic conductivity> composition so that [lithium ion]/[an oxyethylene unit] may become equivalent ratio 0.05, it heated at 100 degrees C under reduced pressure of 400Pa for 3 hours, THF was removed completely, and the solid electrolyte was created. This solid electrolyte was fabricated by the stainless steel electrode with a heat press at the style, and the cell for measurement was created after cooling using this. The impedance was measured for the created cell for measurement using the frequency response analyzer (SI[ by the solar TRON company ]- 1287) by the complex impedance measurement method (alternating current amplitude voltage 1V, the frequency band of 1Hz - 2MHz of an alternating current, temperature of 30 degrees C), and ionic conductivity was computed. The result was shown in Table 1.

[0015] The polysiloxane bridge formation object was compounded completely like creation of the film for elastic-modulus measurement, and the example 1 of example 2 <measurement> composition, and some solutions were isolated preparatively after the reaction end on the petri dish made from the poly ethylene fluoride with a diameter of 30mm. The low-boiling part was removed decompressing this gradually so that air bubbles may not enter. It dried at 130 degrees C under reduced pressure of 400 morePa, and the film for measurement was created. The examination side with a thickness [ of 2mm ] x diameter of 5mm was started from the created film, and the elastic modulus in a room temperature was measured according to the procedure of following <GAI ID=0001>\*\* - \*\* using the heat mechanical analysis equipment TMA100 made from SEIKO Electronic industry. \*\* Set the film created to TMA100, and where load 98mN is added, measure the initial length of a sample. \*\* Measure a TMA amplitude in the load amplitude of 98mN\*\*19.6mN (frequency of 0.01Hz). \*\* Substitute end-of-the-probe area (5.3mm<sup>2</sup>), load amplitude (39.2mN), the initial length of a sample, and a TMA amplitude for the definition formula (definition formula 1) of an elastic modulus, and compute an elastic modulus. The obtained elastic modulus was shown in Table 2.

(Elastic modulus) = {(load amplitude)/(end-of-the-probe area)} / {(TMA amplitude)/(the initial

length of a sample)} (definition formula 1)

[0016] Toluene 50mL [ which dehydrated one or more evenings by molecular-sieve 3A ], and triethylene-glycol allyl-compound methyl-ether 16.92g, triethylene-glycol diaryl ether 4.14g, and platinum divinyl tetramethyl disiloxane were put into the four example of comparison composition 1 <composition of polysiloxane graft bridge formation object> 200mL(s) mouth flask so that it might be set to 50 ppm to Si-H in poly methyl hydronalium siloxane 5.00g of average molecular weight 1700, and heating stirring was carried out at 80 degrees C under the N2 seal for 1 hour. Subsequently, the solution which diluted the aforementioned poly methyl hydronalium siloxane 5.00g with dehydration toluene 10mL was dropped. Heating stirring was then continued for 1 hour, after checking that the peak of Si-H has disappeared by IR, the low-boiling part was removed at 130 degrees C under reduced pressure of 400Pa, and the polysiloxane graft bridge formation object of a brown transparent solid-state was acquired almost quantitatively.

[0017] After mixing 0.0115g LiClO<sub>4</sub> uniformly in THF to 0.2g of polysiloxane graft bridge formation objects of the example 1 <creation [ of a solid electrolyte and the cell for measurement ], and measurement of ionic conductivity> above of comparison so that [lithium ion]/[an oxyethylene unit] may become equivalent ratio 0.05, it heated at 100 degrees C under reduced pressure of 400Pa for 3 hours, THF was removed completely, and the solid electrolyte was created. This solid electrolyte was fabricated by the stainless steel electrode with a heat press at the style, and the cell for measurement was created after cooling using this. The impedance was measured for the created cell for measurement using the frequency response analyzer (SI[ by the solar TRON company ]- 1287) by the complex impedance measurement method (alternating current amplitude voltage 1V, the frequency band of 1Hz - 2MHz of an alternating current, temperature of 30 degrees C), and ionic conductivity was computed. The result was shown in Table 1.

[0018] The polysiloxane graft bridge formation object was compounded completely like creation of the film for elastic-modulus measurement, and the example 1 of example of comparison 2 <measurement> comparison composition, and some solutions were isolated preparatively after the reaction end on the petri dish made from the poly ethylene fluoride with a diameter of 30mm. The low-boiling part was removed decompressing this gradually so that air bubbles may not enter. It dried at 130 degrees C under reduced pressure of 400 morePa, and the film for measurement was created. The examination side with a thickness [ of 2mm ] x diameter of 5mm was started from the created film, and the elastic modulus in a room temperature was measured in the same procedure as an example 2 using the heat mechanical analysis equipment TMA100 made from SEIKO Electronic industry. The obtained elastic modulus was shown in Table 2.

[0019]

[Table 1]

<表 1>

	実施例 1	比較例 1
イオン伝導度(S/cm)	$1.8 \times 10^{-5}$	$1.8 \times 10^{-5}$

[Table 2]

<表 2>

	実施例 2	比較例 2
弾性率(MPa)	0.53	0.20

The result shown in above-mentioned Table 1 and 2 shows having the good mechanical strength, without the polysiloxane bridge formation object of an example lowering ionic conductivity compared with the polysiloxane bridge formation object of the example of comparison.

[0020]

[Effect of the Invention] By this invention, also in a non-solvent system, it has high ionic conductivity, and had the further excellent mechanical strength, and it became possible to obtain a stable solid polymer electrolyte also electrochemically and thermally.



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TECHNICAL FIELD

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[The technical field to which invention belongs] this invention is the solid polymer electrolyte which can be used for a primary cell, a rechargeable battery, a capacitor, etc., and relates to the high ion conductivity and the solid polymer electrolyte with a good mechanical strength which are obtained using the polysiloxane bridge formation object acquired by [ of the polysiloxane and polyoxyethylene unit which have 3 functionality silicon ] combining, and this.

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PRIOR ART

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[Description of the Prior Art] The solid polymer electrolyte attracts attention as a material which realizes thin-shape-izing of a cell, spill liquid[ no ]-izing, and heat-resistant improvement. Which stable property is demanded stably and thermally electrochemically, and promising \*\* of the ether system polymer, such as a polyethylene oxide, is conventionally carried out in respect of the motile height of the polymer by a glass transition temperature being low, and lithium ion solubility as a material with a mechanical strength strong against a solid polymer electrolyte with high ionic conductivity with which are satisfied of these properties. Moreover, many methods of graft-izing are used for the low polymer of a glass transition temperature as a means for moving ion efficiently.

[0003] Since the glass transition temperature is low, especially as for the solid polymer electrolyte which used the polysiloxane for the principal chain, research by combination with ether system polymer is made briskly. Furthermore, since the binding energy is quite larger than a carbon-carbon to carbon bond and siloxane combination which makes the skeleton of a polysiloxane has it, it is useful as a material of a solid polymer electrolyte. [ electrochemically stable ] The solid polymer electrolyte which becomes JP,4-56002,A from the graft copolymer of a polysiloxane, a polyether glycol, and the polyether glycol monomethyl ether is indicated, and the solid polymer electrolyte which consists of a copolymer of the organopolysiloxane which has a carboxyl group content hydrocarbon group in JP,7-233243,A, and the polyoxyalkylene which has an epoxy group is indicated.

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## EFFECT OF THE INVENTION

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[Effect of the Invention] By this invention, also in a non-solvent system, it has high ionic conductivity, and had the further excellent mechanical strength, and it became possible to obtain a stable solid polymer electrolyte also electrochemically and thermally.

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TECHNICAL PROBLEM

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[Problem(s) to be Solved by the Invention] However, even if it is which [ using the polysiloxane ] case, sufficient ionic conductivity is not obtained and has not resulted in utilization. And if improvement in ionic conductivity is aimed at using this method, the trouble of reducing a mechanical strength and membrane formation nature conversely is held. this invention has high ionic conductivity also in a non-solvent system, has the further excellent mechanical strength, and aims at offering a stable solid polymer electrolyte also electrochemically and thermally.

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MEANS

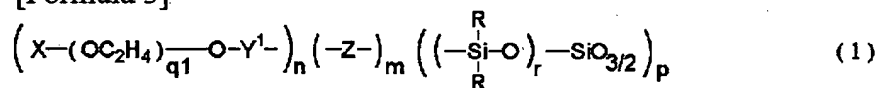
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[Means for Solving the Problem] This invention persons by using the polysiloxane bridge formation object which consists of a polysiloxane which has 3 functionality silicon, and a compound which has a polyoxyethylene unit as a result of repeating research paying attention to a siloxane A mechanical property improves sharply, maintaining the stability of siloxane combination, and moreover the ionic conductivity in a non-solvent system It found out that the graft copolymer of the polysiloxane of a straight chain and the organic macromolecule which has a polyoxyethylene unit, and the solid polymer electrolyte which is more than equivalent were obtained, and this invention was attained. That is, this invention has the following composition.

[0006] (1) The polysiloxane bridge formation object characterized by combining the structural unit which consists of a polysiloxane which has 3 functionality silicon, and the structural unit which consists of a compound which repeats oxyethylene and it has as a unit.

(2) A polysiloxane bridge formation object given in the aforementioned (1) term characterized by being the polymer of the composition as which a polysiloxane bridge formation object is expressed in a formula (1).

[Formula 3]



(X is the alkyl of carbon numbers 1-6 among a formula, Y1 is the alkylene of carbon numbers 2-20, Z is a basis which has the alkylene of carbon numbers 2-20 in both ends, R is the alkyl of carbon numbers 1-6, 0 or 1p of one or more integers and r is the integer of 4-60, and q1, n, and m are n+2 m=p independently, respectively.)

(3) A polysiloxane bridge formation object given in the aforementioned (2) term characterized by Z being the basis expressed with a formula (2).

[Formula 4]



(As for Y2 and Y3, the alkylene of carbon numbers 2-20 and q2 are one or more integers independently among a formula, respectively.)

(4) A polysiloxane bridge formation object given in the aforementioned (3) term characterized by q2 being 3-15.

(5) A polysiloxane bridge formation object given in any 1 term of the aforementioned (2) - (4) term characterized by adjusting the relation between n and m so that  $2m/(n+2m)$  may be 0.2-0.6.

(6) A polysiloxane bridge formation object given in any 1 term of the aforementioned (2) - (5) term characterized by r being 0.

(7) A polysiloxane bridge formation object given in any 1 term of the aforementioned (2) - (5) term characterized by r being 1.

(8) The solid polymer electrolyte which becomes any 1 term of the aforementioned (1) - (7) term from the polysiloxane bridge formation object and lithium salt of a publication.

[0007]

[Embodiments of the Invention] The polysiloxane bridge formation object in this invention is a

polysiloxane bridge formation object which makes it come to join together the polysiloxane which has 3 functionality silicon, and the compound which repeats oxyethylene and it has as a unit. The polysiloxane which has 3 functionality silicon and has an Si-H basis at the end as a synthesis method of this polysiloxane bridge formation object, for example, the compound which has a double bond in both ends, and the compound which has a double bond at the piece end and contains a polyoxyethylene unit in it are mixed in the solvent which can dissolve these compounds, and there is the method of supplying a catalyst to this and carrying out a polyaddition by the hydrosilylation reaction.

[0008] The product marketed may be used for the polysiloxane which has 3 functionality silicon and has an Si-H basis, and it may compound it using what conventionally well-known means. The compound which there is silsesquioxane expressed with general formula  $\text{Si}_2\text{nH}_2\text{nO}_3\text{n}$  (the thing of 4 or 5 is known for n.) as an example known well, and replaced a part or all of an Si-H basis of these compounds by 2-H Si-O-SiR (R shows the alkyl group of carbon numbers 1-6.) can be mentioned.

[0009] Next, the compound which has a double bond in both ends works as a connection machine for combining the polysiloxanes which have 3 functionality silicon, and polyethylene-glycol diaryl ether and end vinyl poly dimethylsiloxane are mentioned. Moreover, as a compound which has a double bond at the piece end and contains a polyoxyethylene unit in it, a triethylene-glycol allyl-compound methyl ether, the triethylene-glycol methyl vinyl ether, triethylene-glycol allyl-compound ethyl ether, a polyethylene-glycol allyl-compound methyl ether, etc. are mentioned, for example.

[0010] As an organic solvent used for a hydrosilylation reaction, toluene, a tetrahydrofuran (THF), etc. are mentioned, for example. Moreover, as a hydrosilylation catalyst, a platinum compound, a ruthenium compound, and a rhodium compound are mentioned, for example.

[0011] The solid polymer electrolyte of this invention blends lithium salt with the above-mentioned polysiloxane bridge formation object. Although especially the combination method of lithium salt is not limited, lithium salt is uniformly mixed with a polysiloxane bridge formation object in an organic solvent, and the method of removing an organic solvent completely under reduced pressure and heating is mentioned after that, for example. Although it will not be limited as an organic solvent used especially if it is dissolution possible about lithium salt, THF, a dimethylformamide, an acetone, etc. are mentioned, for example.

[0012] As lithium salt, as long as it is usually used for a solid polymer electrolyte, you may be what lithium salt. For example, LiF, LiCl, LiBr, LiI, LiClO<sub>4</sub>, LiBF<sub>4</sub>, LiPF<sub>6</sub> and LiAsF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub> and LiN(C<sub>2</sub>F<sub>5</sub>SO<sub>2</sub>)<sub>2</sub>, and LiC(CF<sub>3</sub>SO<sub>2</sub>)<sub>3</sub> grade can be mentioned. What is necessary is to choose at least one or more sorts of lithium salt from these, and just to use.

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## \* NOTICES \*

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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**EXAMPLE**


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[Example] Although this invention is further explained to a detail based on an example, this invention is not limited to these examples.

Toluene 15mL [ which dehydrated one or more evenings by molecular-sieve 3A in the four synthetic example 1 <composition 1 of polysiloxane bridge formation object> 100mL(s) mouth flask ], and triethylene-glycol allyl-compound methyl-ether 0.803g, polyethylene-glycol diaryl ether 0.748g of average molecular weight 300 and 1, 3, 5, 7, 9, 11 and 13, and 15-OKUTAKISU (dimethyl siloxy) PENTA cyclo [9. 5.1.13, 9.15, 15.17, and 13] OKUTA siloxane 1.00g. It put in and heating stirring was carried out at 80 degrees C under the N<sub>2</sub> seal for 1 hour. Subsequently, 50 ppm of platinum divinyl tetramethyl disiloxane were supplied to Si-H, and heating stirring was continued as it was for 3 hours. After checking that the peak of Si-H has disappeared by IR, the low-boiling part was removed at 120 degrees C under reduced pressure of 400Pa, and the polysiloxane bridge formation object of a brown transparent solid-state was acquired almost quantitatively. The acquired polysiloxane bridge formation object is set to PS1.

[0014] After mixing uniformly 45.98x10 to 3 g LiClO in THF to 0.1 g of PS1 obtained in the example 1 of example 1 <measurement of creation [ of a solid electrolyte and the cell for measurement ], and ionic conductivity> composition so that [lithium ion]/[an oxyethylene unit] may become equivalent ratio 0.05, it heated at 100 degrees C under reduced pressure of 400Pa for 3 hours, THF was removed completely, and the solid electrolyte was created. This solid electrolyte was fabricated by the stainless steel electrode with a heat press at the style, and the cell for measurement was created after cooling using this. The impedance was measured for the created cell for measurement using the frequency response analyzer (SI[ by the solar TRON company ]- 1287) by the complex impedance measurement method (alternating current amplitude voltage 1V, the frequency band of 1Hz - 2MHz of an alternating current, temperature of 30 degrees C), and ionic conductivity was computed. The result was shown in Table 1.

[0015] The polysiloxane bridge formation object was compounded completely like creation of the film for elastic-modulus measurement, and the example 1 of example 2 <measurement> composition, and some solutions were isolated preparatively after the reaction end on the petri dish made from the poly ethylene fluoride with a diameter of 30mm. The low-boiling part was removed decompressing this gradually so that air bubbles may not enter. It dried at 130 degrees C under reduced pressure of 400 morePa, and the film for measurement was created. The examination side with a thickness [ of 2mm ] x diameter of 5mm was started from the created film, and the elastic modulus in a room temperature was measured according to the procedure of following \*\* - \*\* using the heat mechanical analysis equipment TMA100 made from SEIKO Electronic industry. \*\* Set the film created to TMA100, and where load 98mN is added, measure the initial length of a sample. \*\* Measure a TMA amplitude in the load amplitude of 98mN\*\*19.6mN (frequency of 0.01Hz). \*\* Substitute end-of-the-probe area (5.3mm<sup>2</sup>), load amplitude (39.2mN), the initial length of a sample, and a TMA amplitude for the definition formula (definition formula 1) of an elastic modulus, and compute an elastic modulus. The obtained elastic modulus was shown in Table 2.

(Elastic modulus) = {(load amplitude)/(end-of-the-probe area)} / {(TMA amplitude)/(the initial length of a sample)} (definition formula 1)

[0016] Toluene 50mL [ which dehydrated one or more evenings by molecular-sieve 3A ], and triethylene-glycol allyl-compound methyl-ether 16.92g, triethylene-glycol diaryl ether 4.14g, and



platinum divinyl tetramethyl disiloxane were put into the four example of comparison composition 1. <composition of polysiloxane graft bridge formation object> 200mL(s) mouth flask so that it might be set to 50 ppm to Si-H in poly methyl hydronalium siloxane 5.00g of average molecular weight 1700, and heating stirring was carried out at 80 degrees C under the N2 seal for 1 hour. Subsequently, the solution which diluted the aforementioned poly methyl hydronalium siloxane 5.00g with dehydration toluene 10mL was dropped. Heating stirring was then continued for 1 hour, after checking that the peak of Si-H has disappeared by IR, the low-boiling part was removed at 130 degrees C under reduced pressure of 400Pa, and the polysiloxane graft bridge formation object of a brown transparent solid-state was acquired almost quantitatively.

[0017] After mixing 0.0115g LiClO<sub>4</sub> uniformly in THF to 0.2g of polysiloxane graft bridge formation objects of the example 1. <creation [ of a solid electrolyte and the cell for measurement ], and measurement of ionic conductivity> above of comparison so that [lithium ion]/[an oxyethylene unit] may become equivalent ratio 0.05, it heated at 100 degrees C under reduced pressure of 400Pa for 3 hours, THF was removed completely, and the solid electrolyte was created. This solid electrolyte was fabricated by the stainless steel electrode with a heat press at the style, and the cell for measurement was created after cooling using this. The impedance was measured for the created cell for measurement using the frequency response analyzer (SI[ by the solar TRON company ]- 1287) by the complex impedance measurement method (alternating current amplitude voltage 1V, the frequency band of 1Hz - 2MHz of an alternating current, temperature of 30 degrees C), and ionic conductivity was computed. The result was shown in Table 1.

[0018] The polysiloxane graft bridge formation object was compounded completely like creation of the film for elastic-modulus measurement, and the example 1 of example of comparison 2 <measurement> comparison composition, and some solutions were isolated preparatively after the reaction end on the petri dish made from the poly ethylene fluoride with a diameter of 30mm. The low-boiling part was removed decompressing this gradually so that a foam may not enter. It dried at 130 degrees C under reduced pressure of 400 morePa, and the film for measurement was created. The examination side with a thickness [ of 2mm ] x diameter of 5mm was started from the created film, and the elastic modulus in a room temperature was measured in the same procedure as an example 2 using the heat mechanical analysis equipment TMA100 made from SEIKO Electronic industry. The obtained elastic modulus was shown in Table 2.

[0019]

[Table 1]

<表1>

	実施例1	比較例1
イオン伝導度(S/cm)	$1.8 \times 10^{-6}$	$1.8 \times 10^{-5}$

[Table 2]

<表2>

	実施例2	比較例2
弾性率(MPa)	0.53	0.20

The result shown in above-mentioned Table 1 and 2 shows having the good mechanical strength, without the polysiloxane bridge formation object of an example lowering ionic conductivity compared with the polysiloxane bridge formation object of the example of comparison.

[0020]

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